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MISSOURI UNIV-ROLLA DEPT OF PHYSICS

COHERENCE IN PENNING IONIZATION AND FIRST KIND COLLISIONS: VOLU--ETC(U)

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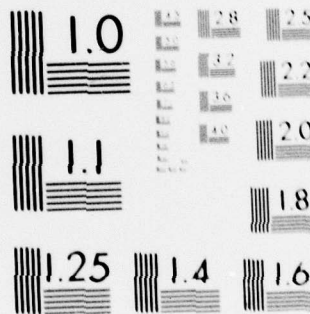
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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) A thermal energy beam of helium atoms in the metastable state is used to coherently excite ions of Sr, Ca, and Ba in a Penning collision. The anisotropic excited state distributions appears as Polarized line emission from the excited ions. The degree of ion alignment may be used to determine the populations of the quasi-molecular states of the ion-atom pair. The coherent emission is also utilized to obtain Hanle signals from which the radiative decay rate of the excited ion is determined. This is the first case in which coherence is produced in a collision process involving Penning		

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ionization. The alignment of the Penning ions is an important new parameter in the description of these reactions.

In an effort to extend such observations to molecular systems a nitrogen molecular metastable beam source was demonstrated. The use of metal vapor targets and the excitation rate of the excited states of Cd and Zn in energy transfer collisions with the N_2^m enable us to determine the vibrational distribution of the $A^3\Sigma$ state of N_2^m in the beam. $N_2(m)$ 3Σ

Energy transfer cross sections and some Penning cross sections were measured in the beam system.

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TECHNICAL REPORT

I Coherence Effects in Penning Ionization

II Volume Recombination Processes at High Pressures

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**Technical Report to the Office of Naval Research
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CONTENTS

	Page
Abstract.....	4
Introduction.....	5
Heavy Particle Interactions.....	5
Energy Loss Processes at High Pressures	7
Progress During This Contract Period.....	9
Publications Issued During This Contract Period.....	11
Other Presentations	12
Progress Report: Summary.....	12
Personnel During This Contract Period.....	14
Advanced Degrees	14
Appendices (Abstracts of Publications).....	15

ABSTRACT

The purpose of the research program is to investigate collisional processes which are important in determining energy balance in discharge plasmas and their afterglows. Two of these processes, which are currently under investigation in our laboratory, are metastable interactions, such as Penning ionization and volume recombination processes.

INTRODUCTION

Heavy Particle Interactions

The general phenomena of energy transfer collisions between neutral atomic and molecular species in a beam and target impurities have been well studied. The amount and type of physical information obtained from studying such collisions has steadily grown with the technology of producing and detecting beams. Energy transfer collisions are those in which either translational or internal energy is transferred between the colliding pair during the collision. This is referred to as collisions of the first and second kind respectively.

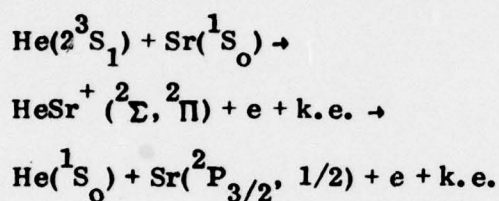
In this research, we examine certain properties of both kinds of collisions for specific collision pairs. The two properties of specific interest are: 1) angular anisotropic distributions and nonstatistical angular momentum state populations in the excited product states of collisions and 2) reaction rates or total cross sections for the collision process.

Knowledge of these properties for a collision pair contributes much to an understanding of the collision dynamics.

Knowledge of the rate at which angular momentum substates are populated in excitation processes yields detailed information about the kinetics of the collision process. Collisional excitation occurring in a beam configuration in general leads to anisotropic excited state distributions, and such distributions manifest themselves as vector and linear polarization of line radiation, orientation and alignment of the excited species, and the nonuniform distribution of collision fragments¹. Such anisotropic distributions have been seen in e-atom², ion-atom³, and ion-beam foil collisions⁴ in which the kinetic energy of

the projectile is the source of the kinetic energy.

In many of the heavy particle collisions, the anisotropic excited states produced are closely associated with the formation of a quasi-molecule during the collision. The observed alignment is then interpreted as a coherent excitation of the quasi-molecular state. Micha et al. demonstrated the utility of the quasi-molecular states in the united atom limit in explaining some of the features of the Penning ionization process⁵. The Penning process is then viewed as a transition between quasi-molecular states; viz:



The process of Penning ionization has been the subject of many experimental and theoretical investigations. Typically, in this process, the excitation energy is transferred to the target atom with the resultant ionization of the target atom. Although most studies have dealt with the total cross section for this process, recently coherent final states have also been observed. In one case, Scheerer has demonstrated in an afterglow experiment that the ions are formed in a coherent state if the metastable atoms are in a coherent state⁶. This has been shown to be a direct consequence of the Wigner spin rule. In a second case, using a beam of metastable helium atoms, Hotop and Niehaus have observed that the electrons ejected in the Penning ionization of argon are in a coherent state⁷. Subsequently, Ebding and Niehaus have shown this to be a general characteristic of the Penning process. In these experiments, the beam serves to define a preferred direction for the system⁸.

The theoretical framework for the Penning ionization process has been presented by

Nakamura⁹ and Miller¹⁰. Ab initio calculations have been performed for the Penning ionization of hydrogen atoms by metastable helium atoms. Semi-empirical calculations have also been made for the ionization of argon by metastable helium atoms. If one assumes with Micha et al. that the electron comes off in a Sigma orbital, the conservation of angular momentum along the interatomic axis permits only the formation of the sigma state of the molecule⁵. One then expects to find that the $m = 0$ levels of the $\text{Sr}^+(^2\text{P}_{3/2})$ ion, referenced to the collision axis, are preferentially populated. If the scattering cross section is nonisotropic, i.e., if the distribution of the orientations of the molecular axes at ionization is anisotropic, then the light emitted by the radiative decay of the $\text{Sr}^+(^2\text{P}_{3/2})$ ions reveals this anisotropy through the polarization of the emitted light. The ion alignment with respect to the beam axis is a coherent state of the ion formed in the Penning process.

Total cross sections for energy transfer collisions are relatively easy to make. Much work on the experimental and theoretical aspects of these measurements has been done in this laboratory and by other workers¹¹. On the other hand, the effects of anisotropic distributions and nonrandom populations are considerably more difficult to observe experimentally. Consequently, much less work has been done in this area. To date, most of the related work involves energetic ion-neutral collisions, but recently some neutral-neutral systems have also been studied¹². The observation of polarized line radiation in a neutral-neutral collision of the second kind, Penning ionization, is reported here for the first time¹³.

Energy Loss Processes at High Pressure

In the afterglow of a high power, the high pressure pulsed discharge, energy loss mechanisms can occur within the context of the collisional-radiative recombination model¹⁴.

Modeling studies of high power laser systems have focussed attention on the lack of

adequate knowledge of many of the rate constants, which must be incorporated into the model. In particular, collisional-radiative and dissociative recombination processes in these systems play a very important role in determining the electron energy distribution. Excited state and ion populations and the gain of these discharge excited mediums depend sensitively on the competition between multistep ionization/excitation by fast electrons and the recombination processes¹⁵.

In more general terms, recombination processes represent loss mechanisms for ions and electrons in high pressure plasmas, such as are present in MHD converters, fusion plasmas, and rocket exhaust plumes.

In the C-R model, the upper energy levels are in Saha equilibrium with the ions and electrons. This implies that their populations will have a Maxwell-Boltzmann distribution. The electron density and electron temperature can then be calculated from the excited state densities according to the Saha equation. And, if the excited states remain in Saha equilibrium as the plasma decays, one can obtain a temporal display of the density and temperature in the afterglow of the pulsed discharge.

In general, it is difficult to obtain a uniform discharge at high buffer gas pressure (> atmosphere). Consequently, we have developed a laser-induced plasma technique in which energy loss mechanisms due to C-R recombination can be observed¹⁶.

This new technique has been applied to the study of volume recombination processes in alkali and alkali-rare gas mixtures. Resonant two-photon ionization of the alkali atom creates a well-defined line source of charge in the vapor cell. The time dependent decay of electron density or optical emission from excited states of the alkali atom are monitored in the afterglow of the fast laser pulse.

PROGRESS DURING THIS CONTRACT PERIOD

The following research accomplishments are described in terms of articles that we have had published or are in the various stages of publishing. The manuscript titles and abstracts along with their current status are listed in the following section on Publications.

Manuscripts A and C listed in the next section describe the results of polarization measurements on the optical radiation produced in two classes of collisions. In the first class, an 800 eV $\text{He}(1^1\text{S}_0)$ atom beam produces excited neutral states in Ca and Sr targets by collisions of the first kind. In the second class, a thermal beam of helium metastable atoms produces excited ions of Ca, Sr, and Ba by collisions of the second kind-Penning ionization. In both cases, the optical radiation from the excited product target states was measured and found to have a net linear polarization with respect to the beam axis. The observation of polarization of line radiation in both classes of collisions is a sensitive measure of the populations of angular momentum states immediately after the collision and of the angular distribution of the collision axes with respect to the beam direction. In manuscript C, a theoretical framework is developed that relates the state populations and the collision axis distribution to the observed polarization in the Penning collisions.

Manuscripts B and D describe the results of lifetime measurements for a neutral level of Ca and Sr and an ion level of Sr. The measurements utilized the Hanle effect or zero field level crossing techniques. The necessary excited state coherence, which in this case results in polarized emission, was produced by the same collisions described above. Such measurements demonstrate the usefulness of polarized, target state emission beyond the understanding of the collision process. The Sr ion level measurement represents the first such measurement in which thermal energy collisions of the second kind were used to pre-

pare the excited state.

Manuscript E describes the results of cross section measurements made for helium metastable atoms incident on Cd and Zn by using the ion collection method. A specially designed oven containing the target vapor was used in order to provide a well-defined target line-density. The only previous measurements of these cross sections were obtained from pulsed stationary afterglow experiments. The measurement of a total quenching cross section adds important information to the knowledge of the specific collision process and serves to complement the excited state alignment measurements.

Manuscript F describes the results of optical emission measurements made on Cd, Zn, and Sr targets excited by excitation transfer from a thermal nitrogen metastable beam. From the variation of the intensity from the target states as a function of the energy of the state, the vibrational temperature of the metastable nitrogen was inferred. This experiment demonstrates a sensitive beam-diagnostic method to measure the vibrational temperature of a thermal, molecular metastable beam. Such a diagnosis is necessary if the alignment measurements described for the atomic beams are to be extended to molecular systems.

In addition to the radiation intensity measurements, linear polarization measurements were made on the emission lines of Cd, Zn, and Sr excited by the nitrogen beam. The measurements were limited to the strongest emission lines and in all cases yielded a null result.

Central to the studies reported here is a reliable and versatile atomic and molecular beam source with a high flux output. As part of this work, such a beam source was developed to produce energetic ground state beams and thermal rare-gas and molecular metastable

beams. The metastable source design is described in manuscript G.

Coherence transfer is the subject of manuscript H. The work on recombination is described in manuscript I. A related laser article is given in J.

PUBLICATIONS ISSUED DURING THIS CONTRACT PERIOD

The following list gives the titles of the manuscripts mentioned above.

- A. "Non-Statistical Excitation of the Magnetic Substates of the Level of Group II Metal Atoms in Collision with 800 eV Helium Atoms", *Physics Letters*, 65A, 215 (1978).
- B. "Hanle Lifetime Measurements of Sr I P and Ca P Levels Excited by a Neutral Beam of Helium Atoms", *Physics Letters*, in press.
- C. "Alignment of Ions in Penning Collisions", *Physical Review*, A20, 1372 (1979).
- D. "The Hanle Effect in Penning Excited Ions", *Journal of Physics B*, 12, L619 (1979).
- E. "Total Penning Ionization Cross Sections of Cd and Zn for Helium Metastable Atoms", *Journal of Chemical Physics*, in press.
- F. "Excitation of Cd, Zn, and Ar by a Beam of Active Nitrogen", *Journal of Chemical Physics*, 71, 2840 (1979).
- G. "High Flux Beam Source of Thermal, Rare-Gas Metastable Atoms", *Journal of Physics E*, in press.
- H. "Spin and Coherence Transfer in Penning Collisions" in Progress in Atomic Spectroscopy B, W. Hanle and H. Kleinpoppen, eds. (Plenum Press, N. Y., 1979) pp. 769-77.

- I. "Laser Initiated Afterglow in a High Pressure K-Kr Mixture", Chemical Physics Letters, 60, 130 (December 1978).
- J. "Injection Locked Dye Laser Pumped by a Xenon Ion Laser", Journal of Quantum Electronics, in press.

OTHER PRESENTATIONS

In addition to the refereed publications indicated here, oral presentations were made at the DEAP and the Gaseous Electronics Conference.

PROGRESS REPORT: SUMMARY

1. A thermal energy, high flux, metastable beam source was designed and constructed. The source provides helium, argon, neon, and molecular nitrogen metastable beams. The beam characterization includes particle detection which yields mean energy, absolute flux, and energy spread and optical detection from the target region, which is used to identify the beam composition and the vibrational distribution in the case of molecular nitrogen.
2. A beam-gas cell arrangement was designed and constructed that permits absolute Penning cross sections to be made. It is particularly useful when channels for metastable quenching are available that involve processes other than Penning ionization. The number of ions can be counted directly, and the metastable loss rate monitored in transmission experiments.
3. Angular anisotropies in excited state distributions have been observed in collisions of both the first and second kind. Such angular anisotropies appear as polarization of

the line radiation produced in the beam collision process. The nonsymmetric excited state distributions had not been observed in second kind collisions prior to this work.

4. The coherent excitation of excited states in these collisions has permitted us to observe Hanle (zero-field Level crossing) signals in some of the Group II metal atoms. These represent the first and only reports of Hanle signals obtained in neutral-neutral beam interactions and represent an extension of the Hanle method for lifetime measurements to new collisional systems.

5. The observed polarization of the optical emission can be related to the alignment produced in the collision process. We have also shown theoretically that the measurement of the ion alignment and a knowledge of the interaction potential can be used to deduce the relative populations of the quasi-molecular states formed during the Penning collision.

PERSONNEL DURING THE CONTRACT PERIOD
(December 1978 to November 1979)

Dr. L. D. Schearer
Dr. W. F. Parks
Dr. Leo Lam, postdoctoral position
Dr. Javed Hussain, postdoctoral position
Mr. David Fahey, graduate student
Mr. Danny Krebs, graduate student
Mr. Howard Cole, graduate student
Mr. Dean Blankenship, graduate student

Advanced Degrees

Mr. Fahey completed the requirements for a Ph.D. degree in October. He has accepted the award of a National Research Council Fellowship to work with Dr. Fehoenfeld at NOAA Laboratories in Boulder, Colorado and has joined the NOAA staff.

APPENDICES

Abstracts of Publications

APPENDIX A

Physics Letters, vol. 65A, no. 3, March 1978

NON-STATISTICAL EXCITATION OF THE MAGNETIC SUBSTATES OF THE 1P_1 LEVEL OF GROUP II METAL ATOMS IN COLLISION WITH 800 eV HELIUM ATOMS

D. W. Fahey and L. D. Schearer

A large polarization has been observed for the emission lines of strontium and calcium excited by a beam of neutral helium atoms with 800 eV lab energy. The measured value of 15% indicates a preferential population of the magnetic substates of the correlated atom states of the target species. Depolarization of the emission in a magnetic field has been observed demonstrating the feasibility of Hanle-type lifetime measurements.

APPENDIX B

Physics Letters, 1979 (in press).

HANLE LIFETIME MEASUREMENTS OF $\text{SrI } ^1\text{P}_1$ AND $\text{CaI } ^1\text{P}_1$ LEVELS EXCITED BY A NEUTRAL BEAM OF 1^1S_0 HELIUM ATOMS

D. W. Fahey, W. F. Parks, and L. D. Schearer

A 0.8 keV $\text{He}(1^1\text{S}_0)$ beam was used to coherently excite the $^1\text{P}_1$ levels of Sr and Ca targets. The coherence appears as an alignment of the excited state with respect to the beam axis. We report here the results of a Hanle measurement, or a zero-field level-crossing experiment, performed on these coherently excited levels. The radiative lifetimes of the $\text{SrI } ^1\text{P}_1$ and $\text{CaI } ^1\text{P}_1$ levels were measured to be 4.7 ns and 5.3 ns, respectively. These values are in good agreement with conventional Hanle measurements.

APPENDIX C

The Physical Review, A, vol. 20, p. 1372, 1979.

ALIGNMENT OF IONS IN PENNING COLLISIONS

D. W. Fahey, L. D. Schearer, and W. F. Parks

We have observed the alignment of the $5p^2P_{3/2}$ state of strontium ions produced in Penning collisions between an unpolarized beam of helium metastable atoms and a strontium vapor target. The alignment is shown by a linear polarization of the optical emission from the excited ion. For a 66 meV beam of helium metastable atoms we measure a 3.5% linear polarization of the emission relative to the beam axis. We indicate how the alignment may be used to determine the probabilities for populating the various final quasi-molecular states of the ion-atom pair. The alignment of the Penning ions is an important new parameter in the description of these reactions.

APPENDIX D

Journal of Physics, B, vol. 12, p. L619, 1979.

THE HANLE EFFECT IN PENNING EXCITED IONS

D. W. Fahey, W. F. Parks, and L. D. Schearer

A thermal beam of helium (2^3S_1) metastable atoms was used to coherently excite the $^2P_{3/2}$ levels of Ca, Sr, and Ba in a Penning ionizing collision. The coherent excitation of the ions appears as a linear polarization of the optical emission from the excited ions. The degree of linear polarization is 5.5, 3.5, and 0.5% for Ca, Sr, and Ba, respectively, with the polarization parallel to the beam direction. Hanle effect signals from the $^2P_{3/2}$ level of Sr were observed and the radiative decay rate measured.

APPENDIX E

Journal of Chemical Physics, 1979 (in press).

TOTAL PENNING IONIZATION CROSS-SECTIONS OF Cd AND Zn FOR $\text{He}(2^3\text{S}_1)$ ATOMS

D. W. Fahey, W. F. Parks, and L. D. Schearer

The total Penning ionization cross-sections for Cd and Zn for $\text{He}(2^3\text{S}_1)$ atoms have been measured. The method used was that of Penning ion collection from a vapor cell through which a $\text{He}(2^3\text{S}_1)$ beam was passed. The beam energy was 66 meV with a velocity spread of 45%. The measured cross-sections were 106 and 35 \AA^2 for the Cd and Zn targets, respectively.

APPENDIX F

Journal of Chemical Physics, vol. 71, 2840 (1979).

EXCITATION OF Cd, Zn, AND Sr BY A BEAM OF ACTIVE NITROGEN

D. W. Fahey, W. F. Parks, and L. D. Schearer

Excitation of the electronic levels of Cd, Zn, and Sr is observed when these metal vapors collide with a thermal-energy, active nitrogen beam. The beam is extracted from a glow discharge in pure N_2 . The active beam component is inferred to be vibrationally excited N_2 in the $A^3\Sigma_u^+$ electronic state. The absolute relative intensity of the emission lines in each element was measured. The excitation rates of the Cd and Zn target levels were found to depend exponentially on their energies indicating an effective temperature of approximately 4000°K. We believe that this temperature is related to the vibrational temperature of the $N_2(A^3\Sigma_u^+)$ states that excite Cd and Zn in energy transfer collisions. The excitation rates of the Sr levels did not show an exponential energy dependence, which is a result consistent with $N_2(A^3\Sigma_u^+)$ as the active species. The potential of such an emission study as a sensitive beam diagnostic is noted.

APPENDIX G

Journal of Physics, B, 1979 (in press).

HIGH FLUX BEAM SOURCE OF THERMAL RARE-GAS METASTABLE ATOMS

D. W. Fahey, W. F. Parks, and L. D. Schearer

A high flux beam source has been constructed for the production of helium, neon, and argon metastable atoms. The source is a dc electric discharge maintained in an expanding gas. A metastable flux of 3.5×10^{14} , 1.5×10^{14} , and 7.2×10^{13} atoms/s sr has been achieved with most probable energies of 66, 72, and 74 meV for the helium, neon, and argon sources, respectively. Time of flight measurements showed the widths of the respective velocity distributions to be 45%, 27%, and 27%. The beam composition was determined with the use of particle multipliers and by the observation of the optical excitation produced in certain metal vapor targets.

APPENDIX H

Progress in Atomic Spectroscopy, B, Chap. 18, 1979.

SPIN AND COHERENCE TRANSFER IN PENNING IONIZATION

L. D. Schearer and W. F. Parks

The Penning ionization process has been effectively used as a mechanism for energy transfer from one species to another. Penning processes are special cases of collisions of the second kind in which a portion of the internal energy of the first species is utilized to ionize and excite the second species. This mechanism has been successfully applied in the He-Cd and He-Zn lasers, in which the laser levels are excited states of the Cd or Zn ions which have been populated by the Penning collision between helium metastable atoms and the Cd or Zn ground-state neutral atom. The Penning process has also been used as a convenient and efficient source of ground-state ions. In a recent series of measurements ensembles of ground-state ions of Mg, Ca, Zn, Sr, Cd, and Ba were formed with sufficient densities to perform conventional Hanle experiments. In this way the radiative lifetimes and depolarization cross sections of the first excited $^2P_{3/2}$ states of the ions were determined.

In this article we discuss the phenomenon of spin exchange and coherence transfer in the population of the Zeeman levels of ions by the Penning ionization process.

APPENDIX I

Chemical Physics Letters, vol. 60, no. 1, December 1978

LASER INITIATED AFTERGLOW IN A HIGH PRESSURE K-Kr MIXTURE

L. K. Lam and L. D. Schearer

We ionized potassium atoms in a K-Kr ($[K] = 6 \times 10^{14} \text{ cm}^{-3}$; $[Kr] = 2 \times 10^{19} \text{ cm}^{-3}$) mixture with a 120 μJ , 404 nm dye laser pulse in a stepwise absorption process and monitored the fluorescence spectra as a function of time. We saw a fast ($\lesssim 20 \text{ ns}$) initial component due to electron excitation and a slow decay tail ($\approx 10 \mu\text{s}$) due to electron ion recombination.

APPENDIX J

Institute of Electrical and Electronics Engineers Journal
of Quantum Electronics, 1979 (in press).

INJECTION-LOCKED DYE LASER PUMPED BY A XENON-ION LASER

E. R. Carney, D. W. Fahey, and L. D. Schearer

Injection-locking of a dye laser is reported for a 4-mirror ring-cavity dye laser pumped by a xenon-ion laser. Both a He-Ne laser and tunable CW dye laser were used as the injection sources.